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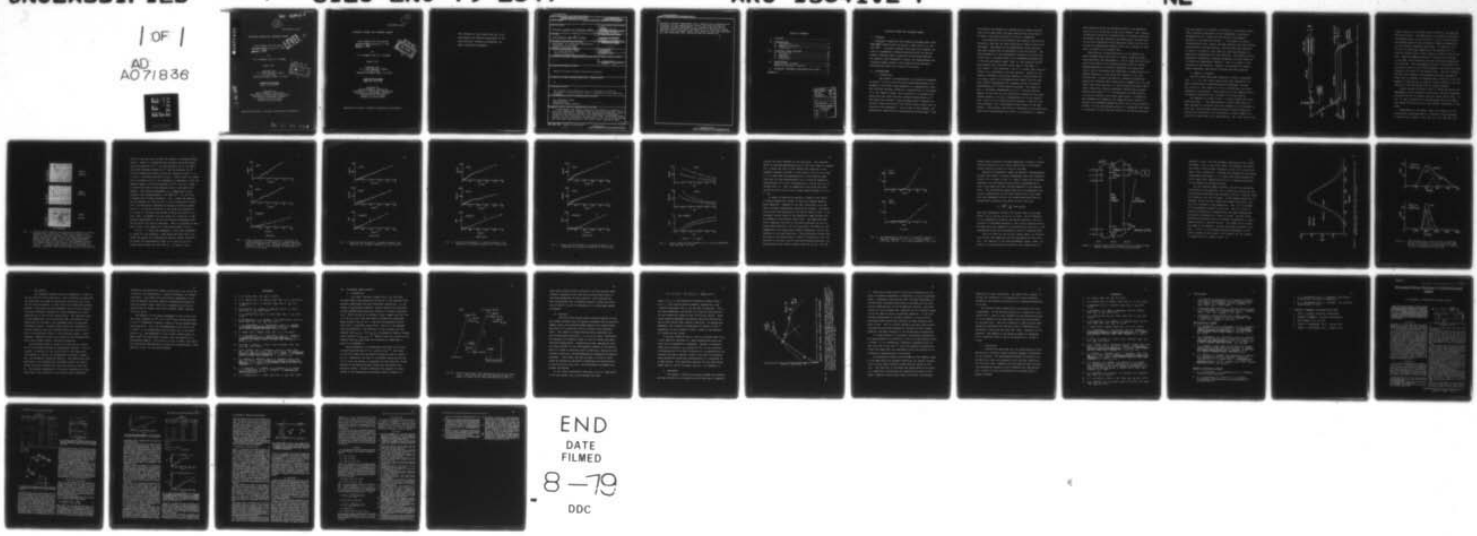

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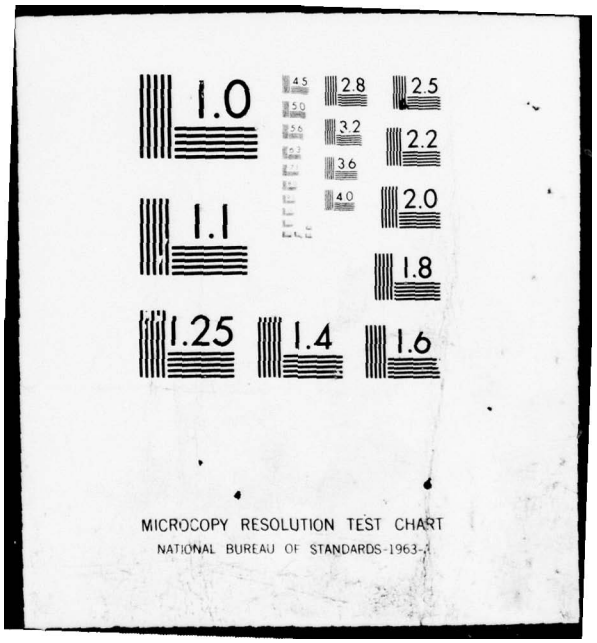
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OPTICALLY PUMPED FAR INFRARED LASERS

Final Report for the Period
~~March 1, 1975~~-January 31, 1979
August 1, 1978-

by

T.A. DeTemple and P.D. Coleman

March 1979

Prepared for
U.S. Army Research Office
P.O. Box 12211
Research Triangle Park, NC 27709

~~DAHC 05-75-G-0099~~
DAAG 29-78-G-0128

Prepared by
Electro-Physics Laboratory
Department of Electrical Engineering
Engineering Experiment Station
University of Illinois
Urbana, Illinois 61801

Approved for public release; distribution unlimited.

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Far infrared lasers
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Experiments and theory regarding the emission processes in optically pumped far infrared laser sources are reported. Superradiance from CH_3F at 496 μm in the homogeneously broadened regime has been identified for the first time. Stimulated Raman emission on pure rotational transitions in D_2O at 50 μm and 66 μm has also been identified for the first time. Generally good

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agreement between experiments and a Maxwell-Bloch approach was obtained for the superradiance study, and good qualitative agreement between experiments and a Maxwell-density matrix approach using three waves interacting in a four-level system was noted for the Raman study; both implying that the processes are now reasonably well characterized and understood.

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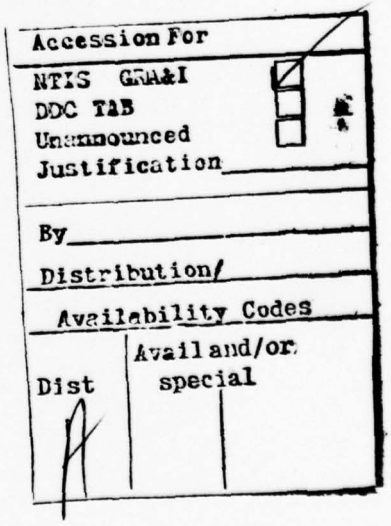

OPTICALLY PUMPED FAR INFRARED LASERS

I. OVERVIEW

This report summarizes the research performed under Grant DAHC 04-75-G-0099 during the period 1 March 1975 to 31 January 1979. The basic research goals were quite simple: the study of fundamental processes associated with the optical pumping technique used to generate far-infrared radiation. Two specific areas addressed in detail are superradiance and stimulated Raman emission, both identified and partially characterized during the course of this study.

II. SUPERRADIANCE

A. Introduction

The concept of superradiance, as originally presented by Dicke,[1] is that of the collective spontaneous decay of an ensemble of two-level atoms prepared in a superposition state. The resultant emission intensity is proportional to the square of N, the number of atoms, and is emitted in a delayed pulse whose delay and width are both inversely proportional to N. The theory has evolved to treat extended media,[2] complete inversion,[3] swept excitation,[4] and various other effects such as relaxation and degeneracy.[5] At the time of the proposal, one experimental observation of superradiance had been made, [6] and

the question was whether our observations on methyl fluoride were of the same effect and what modifications would have to be made in the existing theory[2] to allow a comparison.

The objectives of the proposal were several; and with one exception, they have all been fulfilled at least as well as we had hoped. The details of the superradiant pulse evolution have been studied and are described in the next section. The scaling behavior of the superradiant emission with cross sectional area A, has been found to be intermediate between that of the disk and needle cases,[2] that is, the intensity varies more rapidly than A but more slowly than A^2 (see Ref. 7); with length, L, the variation ($\sim$ as L^3) is faster than that predicted, as is discussed in the next section. Seeding of the sample to determine the initial condition (initial Bloch vector tipping angle) of the Maxwell-Bloch theory[5] has not been done, because reproducible far infrared pulses of intensity approximately 10^{-19} W/cm^2 would be required. The theory of Ref. 5, developed independently by us in a similar version, appears to give not only the qualitative features of the superradiant emission, but also a quantitative fit to our results over a range of cell length and CH_3F pressure, using only one free parameter; this is discussed in more detail in the next section. This analysis has given an indication of the conditions under which superradiance may evolve; in particular, it appears

that population decay and dephasing effects (represented by $T_2=T_1$) are not as serious as was first thought. And, finally, an efficient pulsed far infrared source (15% photon conversion efficiency) has been realized; the efficiency is related only to the finite delay and finite pump pulse width, but the conditions for superradiant emission limit the intensity to less than a kilowatt at the present time.

Contributions in amplification of, or in addition to, those proposed have taken place in several different areas, both theoretical and experimental. One of these was the extension of the theory of the propagationless case[8] to include, phenomenologically, collisional dephasing and energy loss (T_2&T_1) and to investigate the importance of these relative to Doppler dephasing; it was found that Doppler is the less serious case. A search was made for candidate transitions; several far-infrared (FIR) possibilities (pumpable by a CO_2 TEA laser) were found (see Ref. 9), and we identified alkali metal vapor transitions on which subsequent observations of superradiance were made.[10,11,12] The apparatus has been modified to allow for shorter, more stable pump pulses, to further reduce feedback and symmetrize the sample cell with respect to forward- and backward-propagating waves, and to allow sensitive detection of the superradiant pulses on a nanosecond time scale. An analytical solution has been found for the small-area pulse

case and agrees with computer calculations in the proper limits, and an empirical expression for the dependence of the pulse parameters on cell length has been derived from computer solution of the equations of Ref. 4. The effects of varying cross-sectional area, length, and pump duration have been studied experimentally and theoretically, and the pulse shape observed in detail; in addition, the transition from superradiance to swept-gain superradiance has been observed. The transition from homogeneous broadening to Doppler broadening has been found not to affect the superradiant behavior. This is a result of the coherence of the emission. However, the existence of several quasi-independent superradiant processes has been observed.

B. Summary of Research

Earlier results have been reported at several conferences (Ref. 7, 9, 13); the latest results have been reported at the Tenth International Quantum Electronics Conference, Atlanta, May 29-June 1, 1978, paper T-4, and in revised form, are being prepared for publication. These results will be summarized here, and general conclusions will be drawn in the next section.

The apparatus is shown in Fig. 1; certain changes have been made since Ref. 7. The mode quality of the CO_2 TEA laser has been improved by increasing the inner diameter of the intracavity gain cell, and the output energy has been increased threefold, allowing the pump pulse to be truncated to a width (FWHM) of 15 nsec with a peak power still approximately 1 MW. This allows even

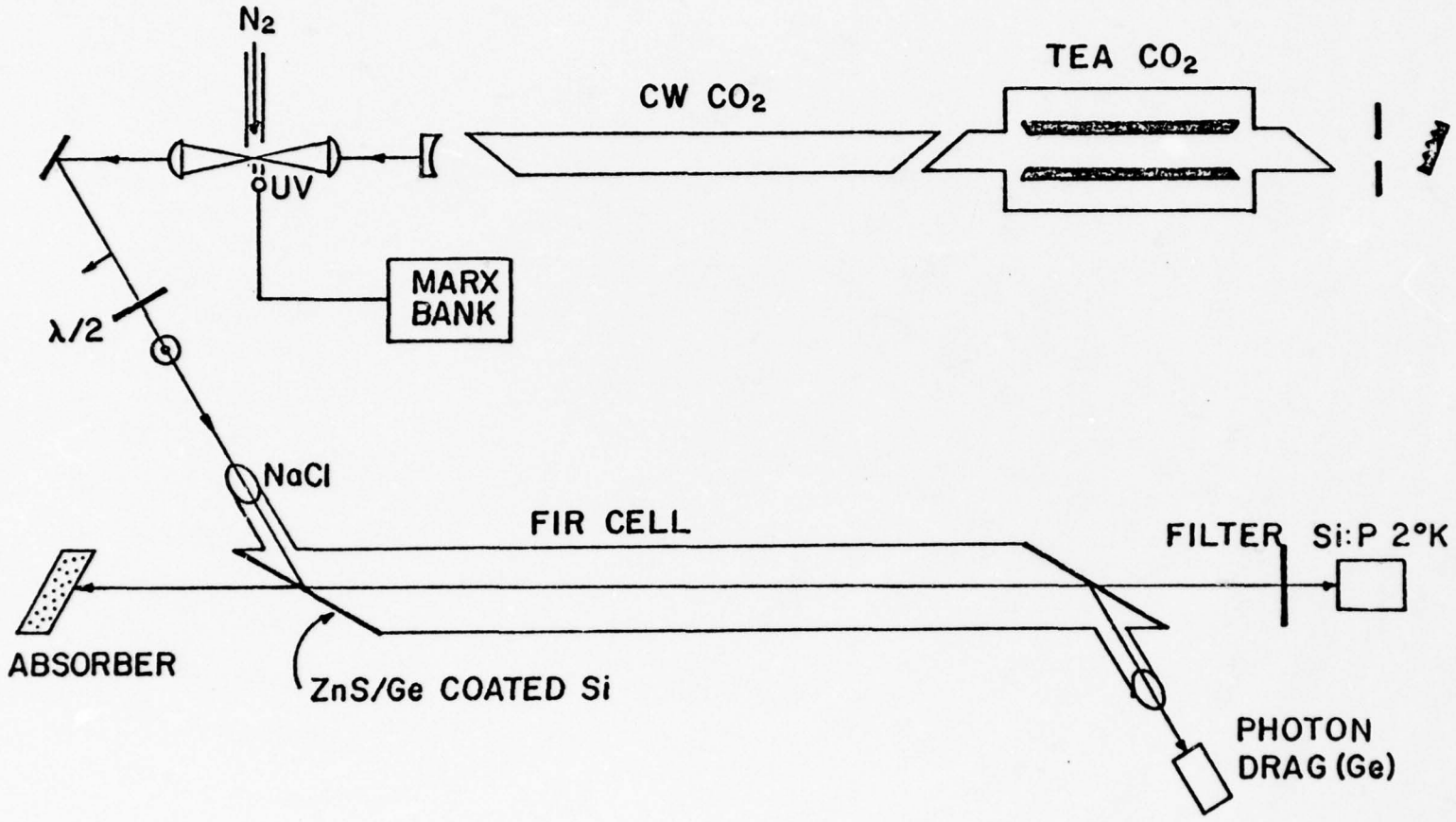

Figure 1. Experimental arrangement showing CO_2 laser, plasma shutter, methyl fluoride cell, and detector.

higher pressure to be reached before overlap of the pump and FIR pulses occurs. The pump pulse is now weakly focused by a long-radius mirror into the FIR cell so that it may traverse the maximum length of cell allowed by laboratory dimensions (10.2 m) without expanding appreciably. In addition, the FIR cell has been made symmetric by the mounting of a dielectric-coated silicon "input coupler" at the far end of the cell, reducing feedback even more, allowing full detection of the pump, and eliminating the need for external separation of the pulses. This arrangement allows for the extension of the range of data to even lower pressures, and allows the measurement of the delay of the backward wave. The Si:P detector has been more carefully calibrated and has been operated at lower bias to decrease its response time to about 2 ns (at the expense of sensitivity) in order to follow faithfully the time behavior of the FIR pulses from the longest samples.

Typical pulses are shown in Fig. 2, where the qualitative dependences of the intensity, pulse width, and delay (measured from pump cutoff) on pressure and cell length can be seen. In Fig. 2c, the fluctuations in the FIR pulses are also evident (the lowest FIR pulse is due to the unusual fluctuation in the pump).

Measurements of the delay, width, and intensity of the superradiant pulses were made as a function of CH_3F pressure at several cell lengths: 168, 229, 351, 473, 656, 838, and

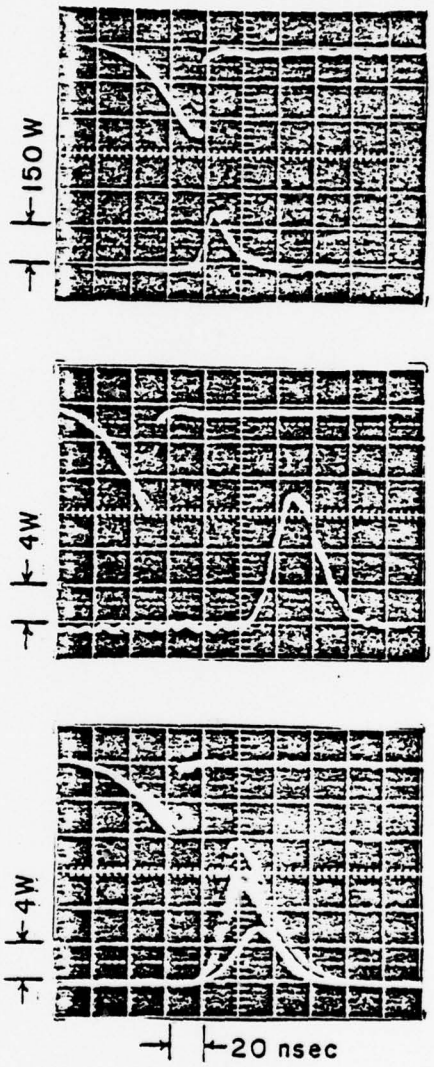

Fig. 2. Typical pulses showing the qualitative effects of varying pressure for a given cell length and of varying cell length at constant pressure. The upper pulse is the CO_2 pump, showing the rapid cutoff; a) nearly-overlapping pulses; vertical scale is 150 W/div (FIR pulse only), horizontal scale 20 nsec/div., b) well-separated pulses at a lower pressure; note change in vertical scale to 4 W/div., c) five pulses at .242 torr in a shorter cell, showing a somewhat larger-than-normal range of fluctuation.

1021 cm; this was done for both the forward- and backward-going pulses. Since it is expected that the delay and width should vary with pressure as p^{-1}, and the intensity as p^2, the delay and width have been plotted vs p^{-1} and the intensity vs p^2 for the forward-going wave at four cell lengths in Figs. 3, 4, 5, and 6. The delay and width in Fig. 3 (168 cm cell) are shown with a straight-line fit; the agreement is reasonable. Note the negative delay at infinite pressure ($p^{-1}=0$); this is a result of the finite width of the pump pulse and is the same at all lengths. The intensity dependence is a combination of two straight lines (approximately), one below p^2 = .01 $torr^2$ and a steeper one at higher pressures. Fig. 4 shows the same behavior observed in a 351 cm cell; in all of these plots (Figs. 3-6), the data points represent an average of several pulses. In Fig. 5, at 656 cm, the break in the slope of intensity vs p^2 is seen to correlate with breaks in delay and width vs p^{-1}; this, then, is assumed to be the result of a variation in the pressure dependences of the two main emission processes which are occurring (k=2 and k=3 emission). This is discussed in more detail later. This behavior is even more evident in Fig. 6.

In Fig. 7 is shown the dependence of the pulse parameters on length for two pressures. Both delay and width show a decrease and appear to be approaching nonzero values; the shortest delays are approximately equal to T_2, and all but the longest pulse widths are less than T_2. In spite of this,

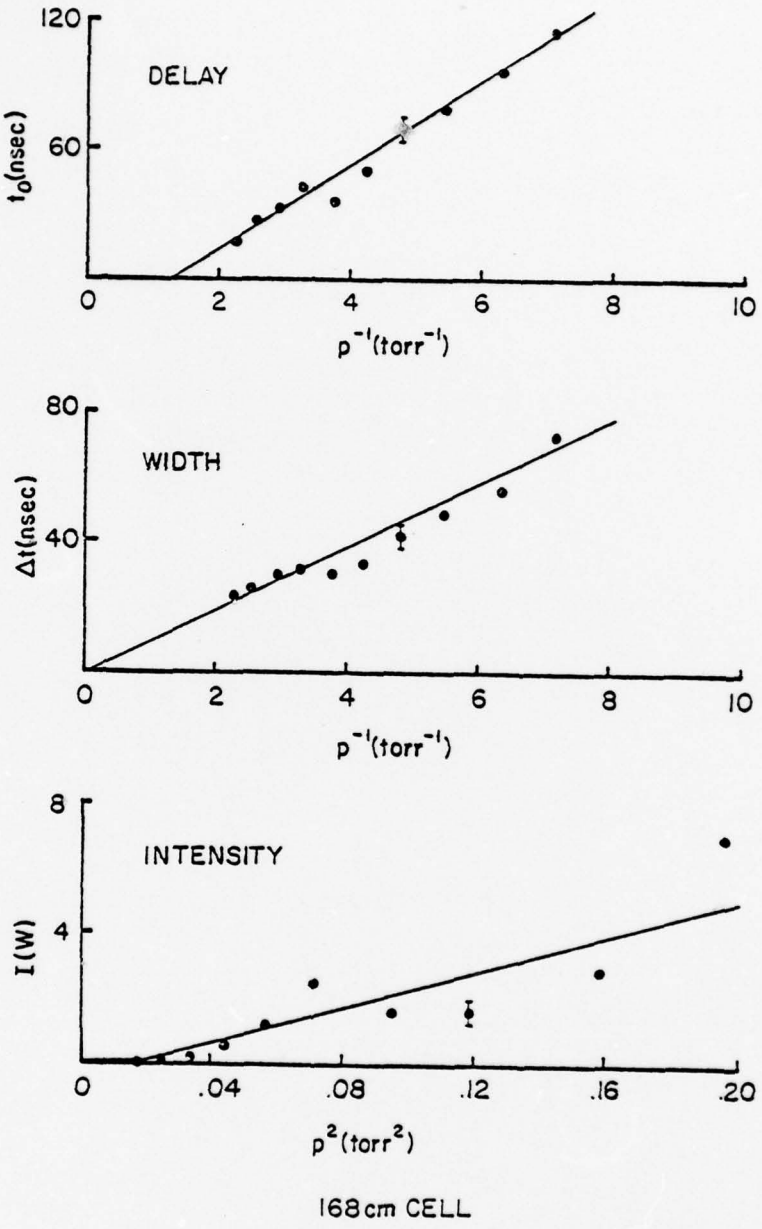

Fig. 3. Delay (measured from pump cutoff to FIR peak) vs. inverse pressure, pulse width vs. inverse pressure, and pulse intensity vs. pressure squared, for a cell length of 168 cm. The intercept of the delay at $p^{-1} = 0$ is -23 nsec.

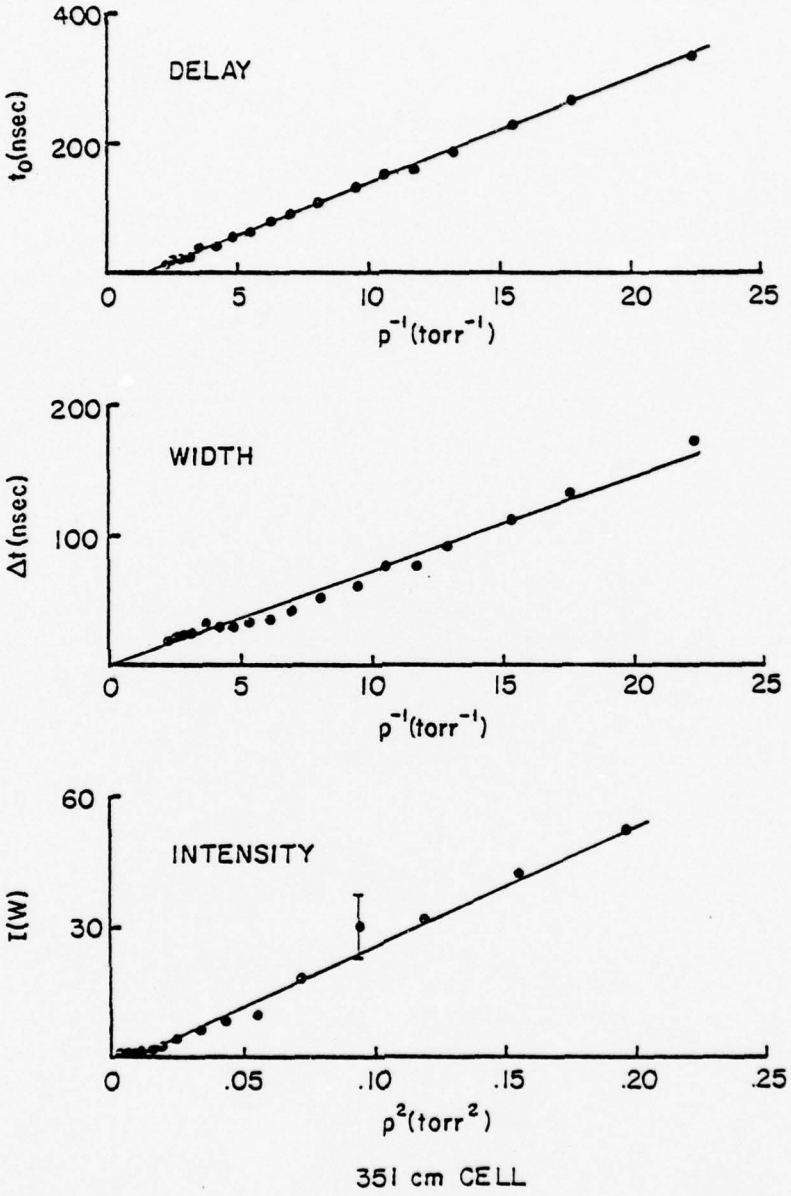

Fig. 4. Delay and pulse width vs. inverse pressure, and intensity vs. pressure squared for a 351 cm cell.

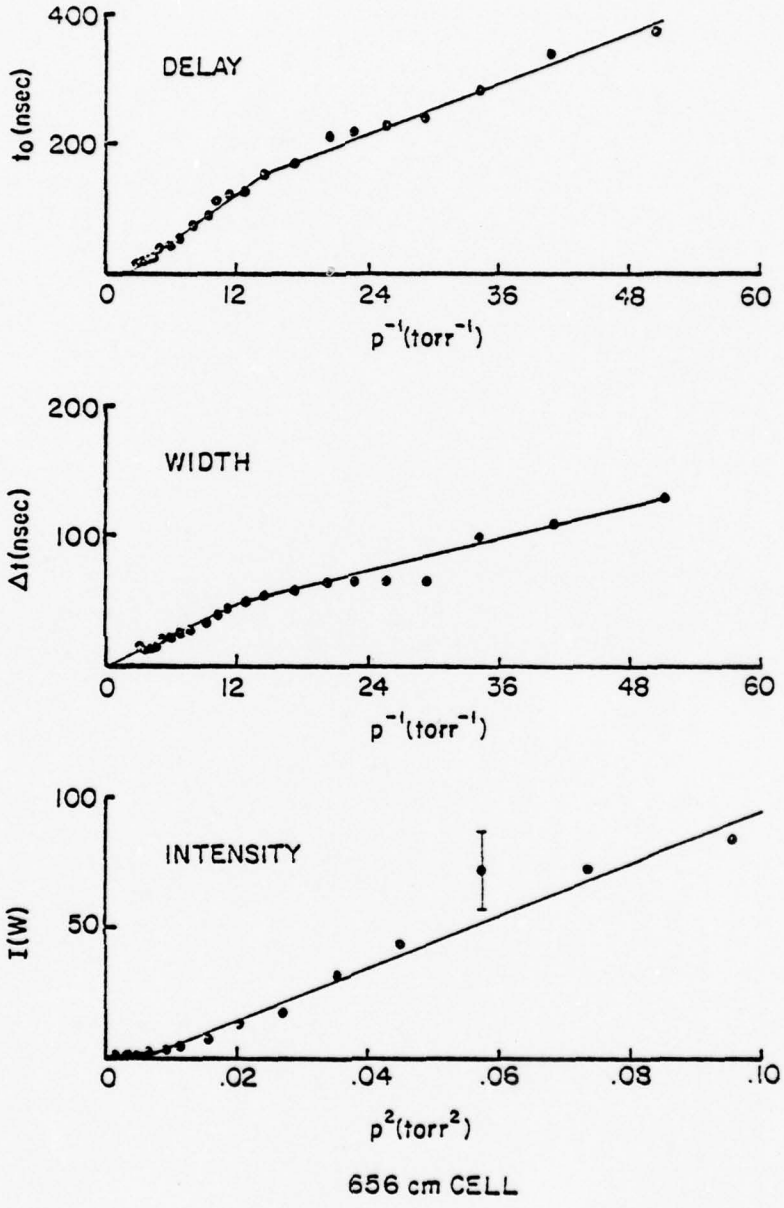

Fig. 5. Delay and pulsewidth vs. inverse pressure, and intensity vs. pressure squared for a 656 cm cell.

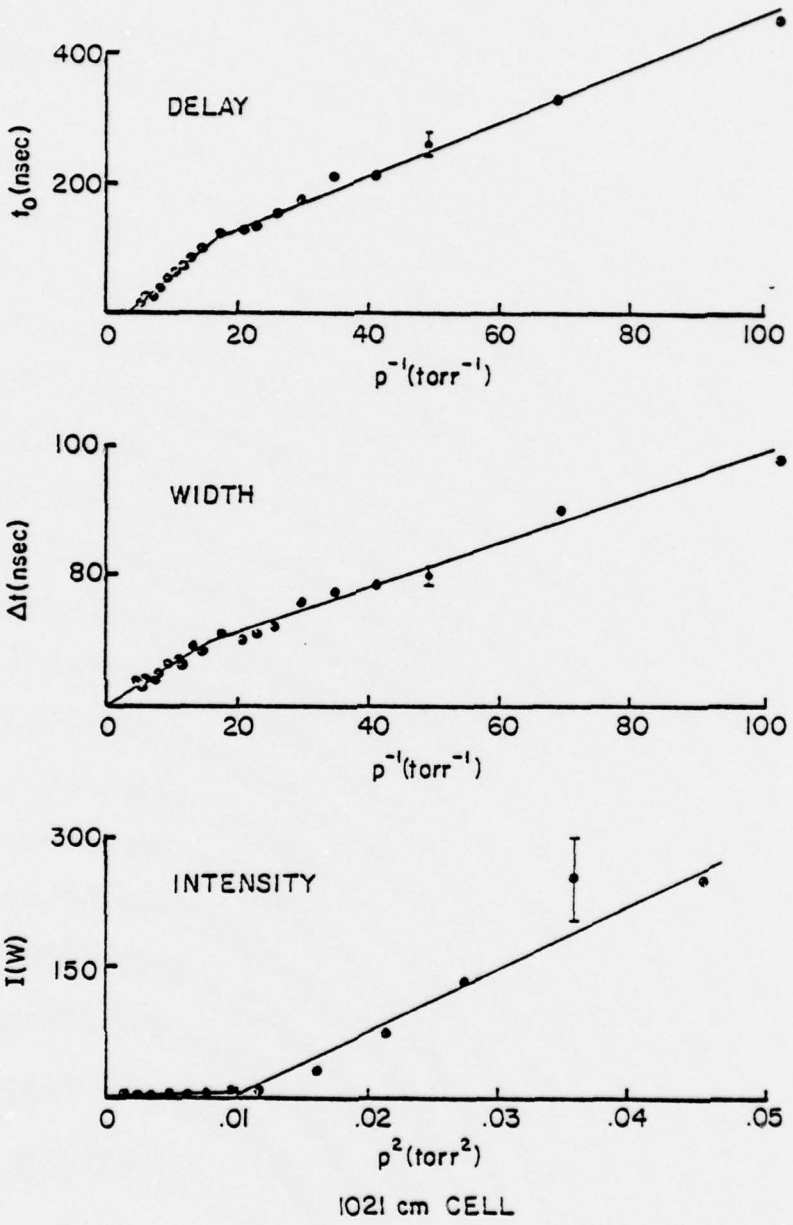

Fig. 6. Delay and pulse width vs. inverse pressure, and intensity vs. pressure squared for a 1021 cm cell.

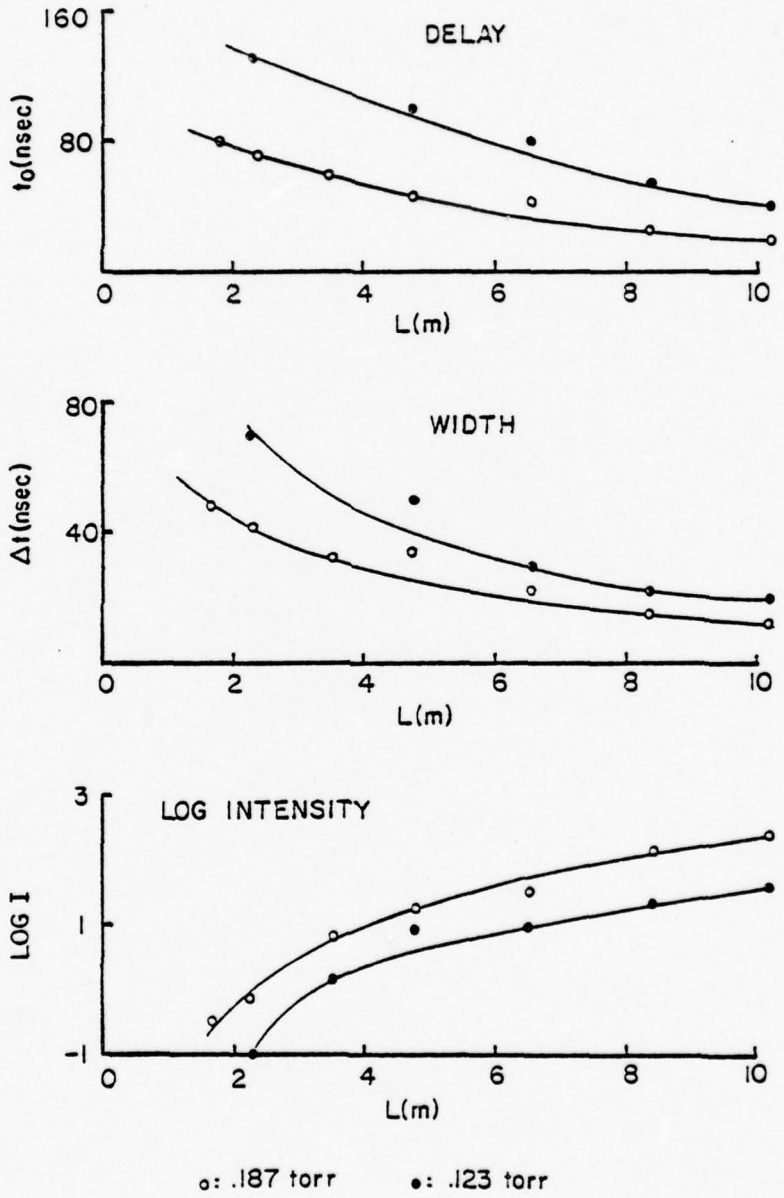

Fig. 7. Delay, width and log intensity vs. L for two pressures, .187 torr and .123 torr.

ringing was never observed in the FIR pulses. The intensity shows an increase approximately as L^3 over this range of lengths. Fig. 8 shows the ratio of the forward FIR intensity to the backward intensity plotted vs p for fixed L and vs L for fixed p; it is seen that the characteristic of the emission goes from that of Dicke superradiance ($R \simeq 1$) to that of swept-gain superradiance ($R >> 1$) as p increases past .10 torr and as L increases past 5 m. This is supported by the delay and width variation with L; both go as a larger negative power of L for $L > 5m$.

In order to explain our results in terms of the theory, a Fortran program was written to solve the complex Maxwell-Bloch equations. Because of run time limitations, only the case of forward superradiant emission was treated. The equations solved were essentially the same as those of Ref. 5, but included a source term for the population (pump process) and a fluctuating polarization source term. The pump term came from a numerical solution of the Bloch equations for the infrared pump transition, and its calculation included the time dependence of the pump pulse, Doppler broadening, and the degeneracy of the pump transition. The polarization source term was taken to be proportional to $[N(t)]^{\frac{1}{2}}$ (statistical) for the duration of the pump, and its fluctuating phase resulted in an emitted pulse that varied slightly from run to run. The FIR

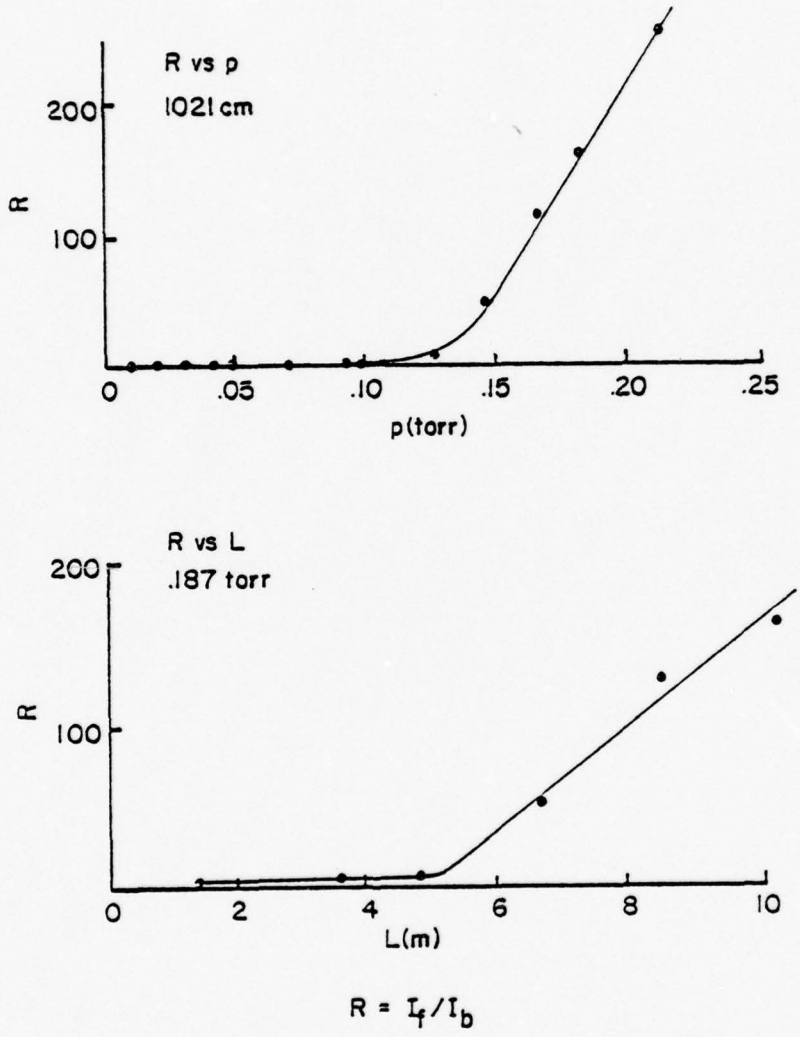

Fig. 8. The dependence of the ratio of forward intensity to backward intensity, R, vs. p at a fixed length (1021 cm), and vs. L at a fixed pressure (.187 torr).

Maxwell-Bloch equations included degeneracy, allowing a fairly accurate precition of the linear polarization of the superradiant emission relative to that of the pump.

Because the absorption (pump) and emission (superradiance) process could occur in several independent ways (for different values of K, as shown in Fig. 9), each of these was calculated separately for the dominant polarization (perpendicular to that of the pump) and their sum was compared to the observed pulse. The polarization souece term can be intrepreted as an initial tipping angle of the Bloch vector, and it was found that good agreement with all our forward-wave data could be established by choosing this angle to be of the form

$$\theta_o = \left(\frac{L_a}{L}\right)^4 \frac{2}{\sqrt{N}} \exp(-T_b/T_R),$$

where the exponential reflects the finite width of the pump pulse,[14] L_a is 900 cm, and T_b is 1.6 nsec. The L^{-4} behavior is not understood at present, but may be related to the contribution of balckbody emission to the initial effective field. The pump pulse was assumed to be saturating and the loss of the superradiant pulse was calculated as that of a Gaussian beam.

With this form for the initial tipping angle, or polarization source, the numerical solution reproduced our data very well. The behavior of the pulse parameters (delay, width, intensity) as the pressure was varied in the program gave our

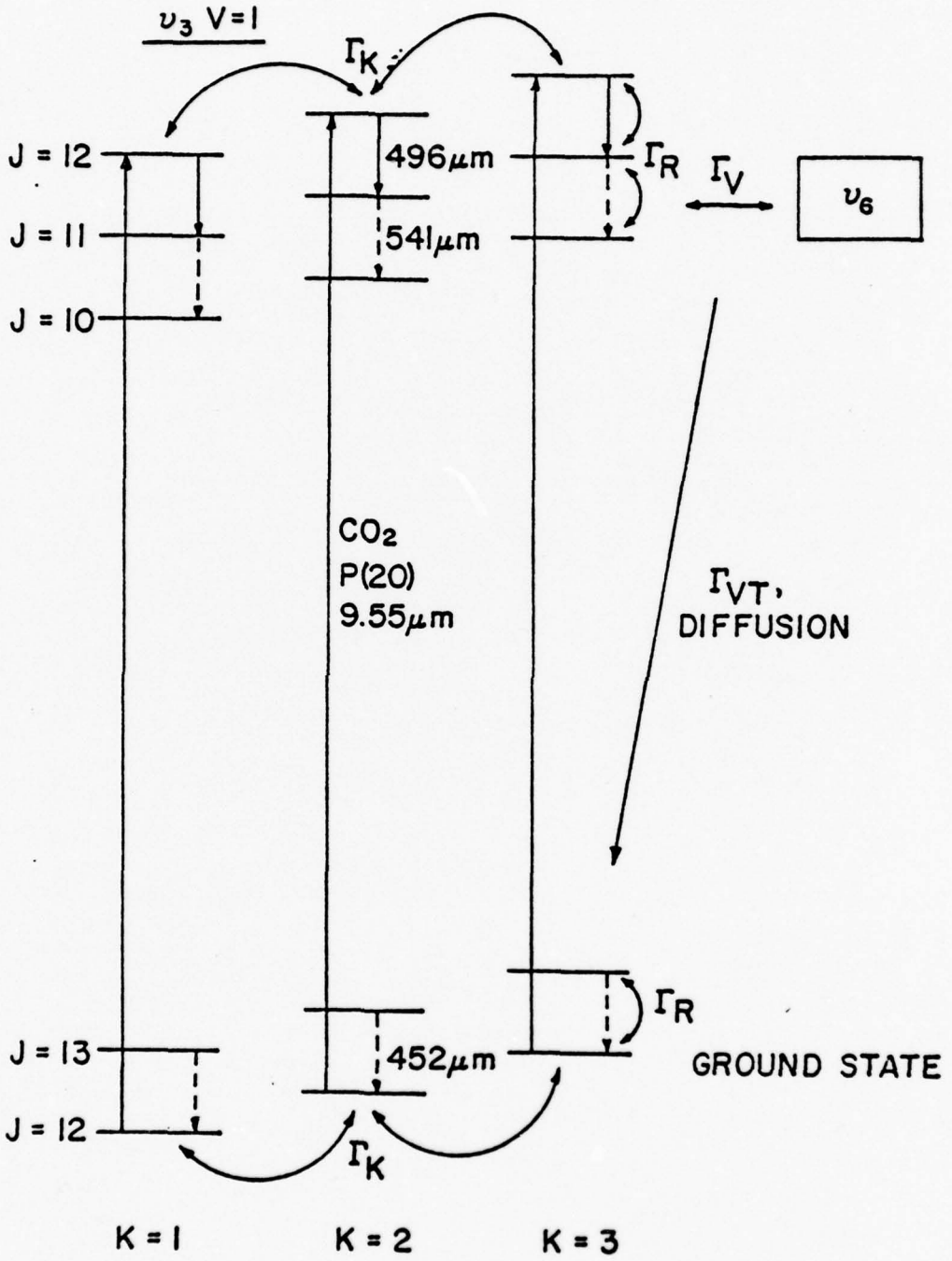

Figure 9. Partial energy level diagram of the ν_3 mode of CH_3F, showing radiative and collisional processes.

observed results, with the agreement improving as cell length increased. Even in the worst cases, the agreement was within the range of shot-to-shot fluctuation (about 10% in width and delay, 20% in intensity). Although the observed and calculated fluctuations were equal, it is likely that those observed are due to pump laser instabilities ad not to quantum fluctuation in the cooperative emission process.

For the shortest sample lengths, only the K=2 transition contributes to the emission; Fig. 10 shows how closely the calculated pulse reproduece that observed, not only in delay, width, and intensity, but also in shape. As the sample length was increased, the K=3 and K=1 transitions became significant, and had to be included in the comparision. Fig. 11 is an example of the agreement obtained in this case. Due to an error in detector calibration at reduced bias, the upper experimental pulse must be reduced by a factor of three. Although these different-K emissions occur at different frequencies, no beats appear because of several factors: the length of the pulses, the speed of the detector, and the transverse intensity variation. In Fig. 11 the different pressure dependence of the three K emissions can be seen; this accounts for the changes in slope which were noted in Figs. 3-6.

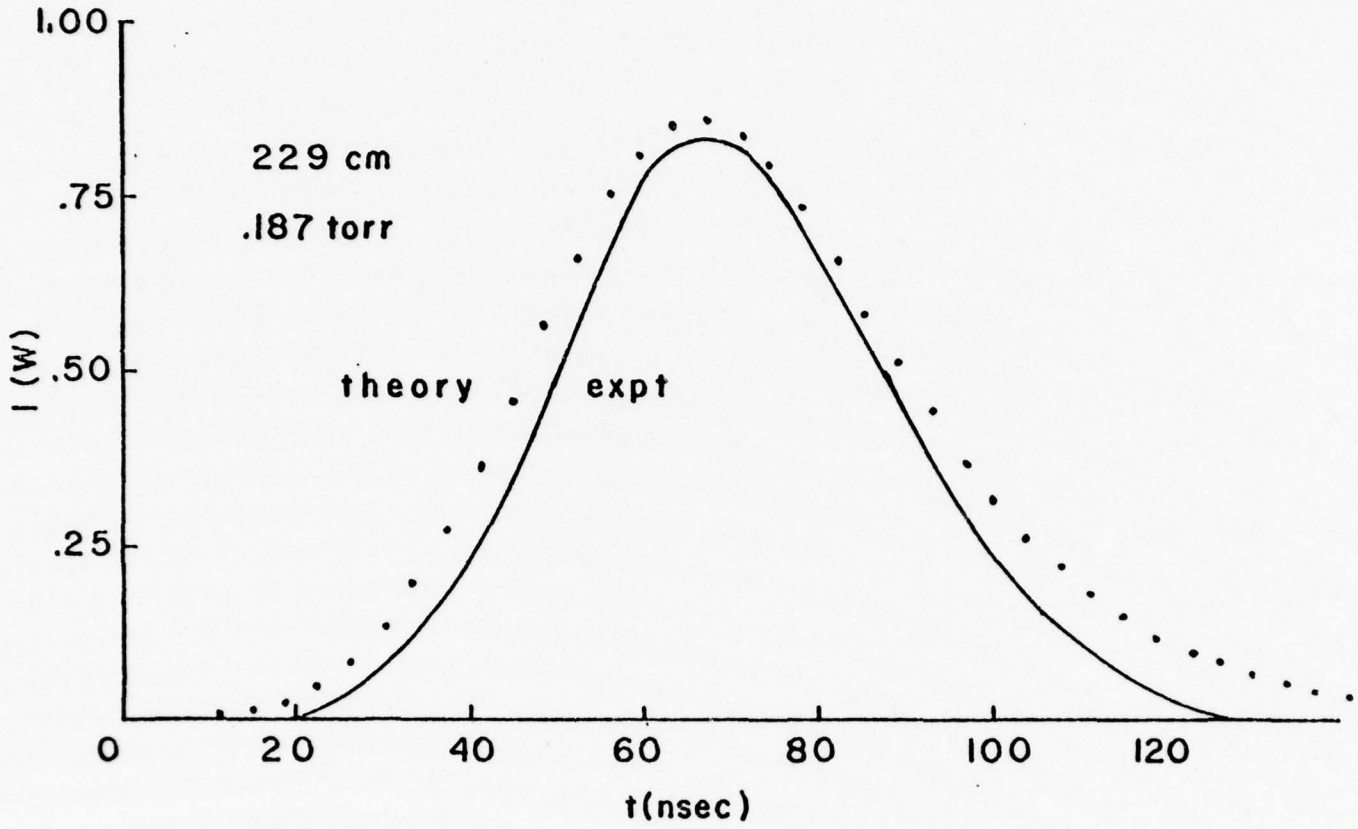

Figure 10. A comparison of the observed (solid line) and calculated (dots) superradiant pulse shape; p = .187 torr, L = 229 cm.

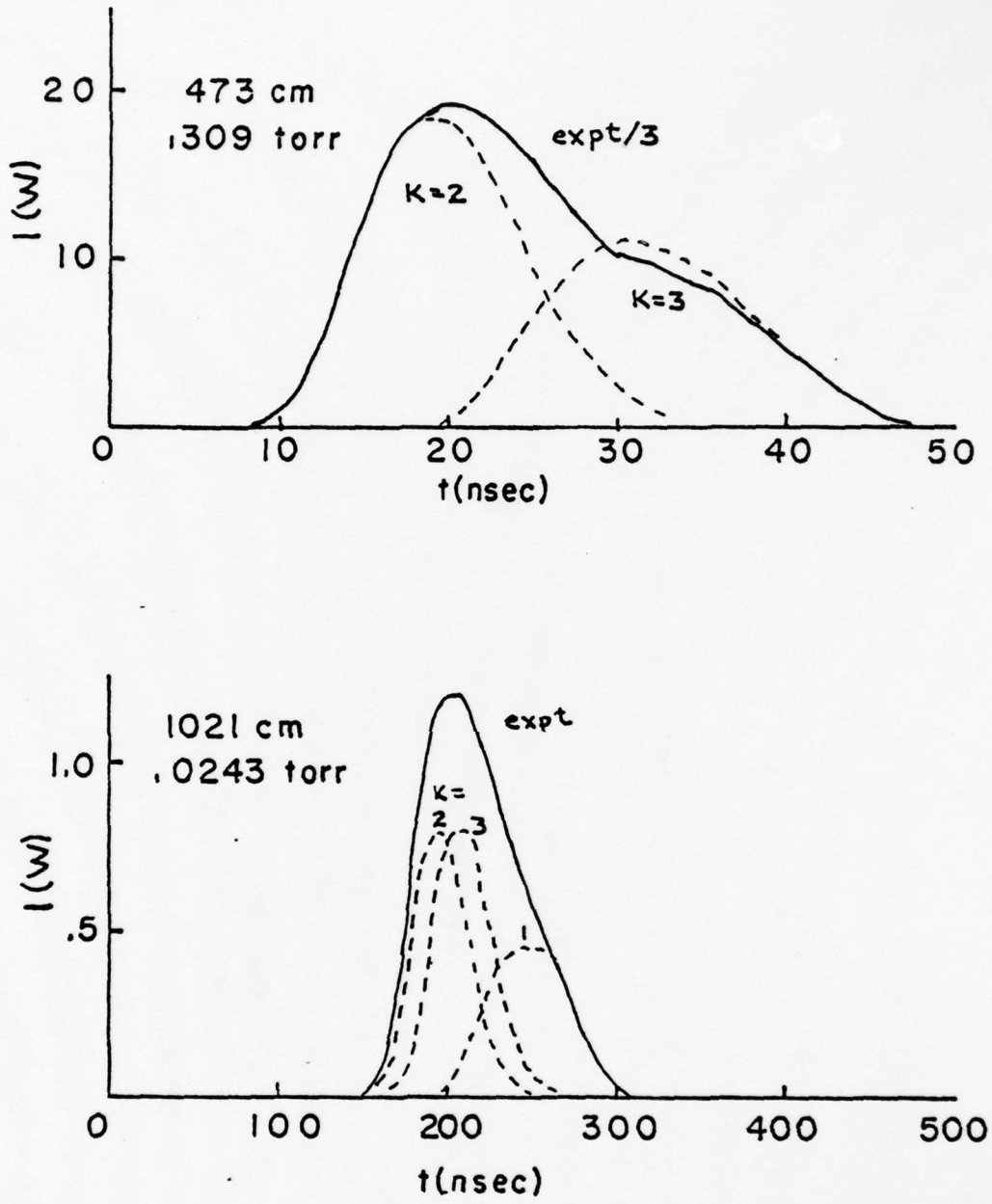

Figure 11. Semiclassical fits (broken lines) to observed pulses (solid lines), in cases where two (top) and three (bottom) K emissions must be combined to give a reproduction of the data.

C. Conclusions

The agreement between theory and experiment is seen to be very good for the forward wave. This claim must be qualified by noting that the sample cross-section and linear field loss are only best estimates, as they could not be measured directly; however, indications are that agreement could also be obtained for other reasonable estimates of these parameters by slight modification of the initial tipping angle. The important point is that degeneracy, the finite-width pump, and several K processes must be included to get agreement over all pressures and sample lengths. Coherent pump effects and transverse intensity variation were not included explicitly and may also have a non-negligible effect on the resulting output. Unfortunately, comparison could not be made with the backward-wave data, which showed several interesting anomalies.

Far-infrared pulses as short as 12 nsec and as intense as 250 W have been produced by superradiant emission. This is limited, at present, by the finite width of the pump pulse, the presence of several (K) superradiant processes, degeneracy, and (probably) transverse effects. The observations have been made in the homogeneously broadened regime, and single pulses have been observed even when the delay was less than T_2. The pressure dependence of the pulse parameters has been explained by the interplay of the several quasi-independent

absorption and emission processes taking place, and not by the transition from homogeneous to Doppler broadening, as thought previously. The length and cross-section dependence of the pulse parameters has shown behavior intermediate between the disk and needle limits, which is in agreement with that expected from our system, which has a Fresnel number slightly less than unity.

The question of the proper dependence of the initial tipping angle (whether as $N^{-\frac{1}{2}}$ or as $(\mu N)^{-\frac{1}{2}}$) has not been settled, as our L^{-4} dependence produces a range of values between these two predictions. Also, the ℓnN-dependence of the delay (ℓnN or $(\ell nN)^2$) is not determinable from our data due to the uncertainty in N. The dependence of the initial tipping angle on pump width[14] has been shown to apply, with the meaning of θ_o interpreted properly. And finally, as Fig. 8 shows, the transition from Dicke superradiance to swept-gain superradiance, first reported in Ref. 13, has been observed.

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III. STIMULATED RAMAN EMISSION

A. Introduction

D_2O vapor, optically pumped with a CO_2 TEA laser, has been under study regarding the nature of far-infrared (FIR) emission associated with pure rotational transitions. In the course of preliminary research, several features of particular interst emerged which have been subjects of further investigation. The 50 μm and 66 μm emission lines, pumped with the 9.66 μm P(32) CO_2 line, have been shown to constitute stimulated Raman emission and to lie $\sim$ 2 GHz from corresponding ground state and ν_2 rotational transitions. This is to be compared with the measured 1.1 GHz detuning of the CO_2 pump from the relevant 000 $6_{61},6_{60}$ → 010 $5_{50},5_{51}$ IR absorption doublet (energy level notation is $J_{K-1,K+1}$).[13] Several other D_2O transition detunings from CO_2 pump lines were measured as summarized in Table II of Appendix I.

Various emission line assignments were made on the basis of recent spectroscopy of the ν_2 band of D_2O.[14] The scheme of the P(32) pump line and observed emission lines is shown in Fig. 12. Neglecting the weak 83 μm cascade transition and the 116 μm line, the system is comprised of three radiation fields present on the dipole-allowed transitions connecting four molecular levels. We have undertaken the analysis of such a system in the homogeneously broadened regime by means of a

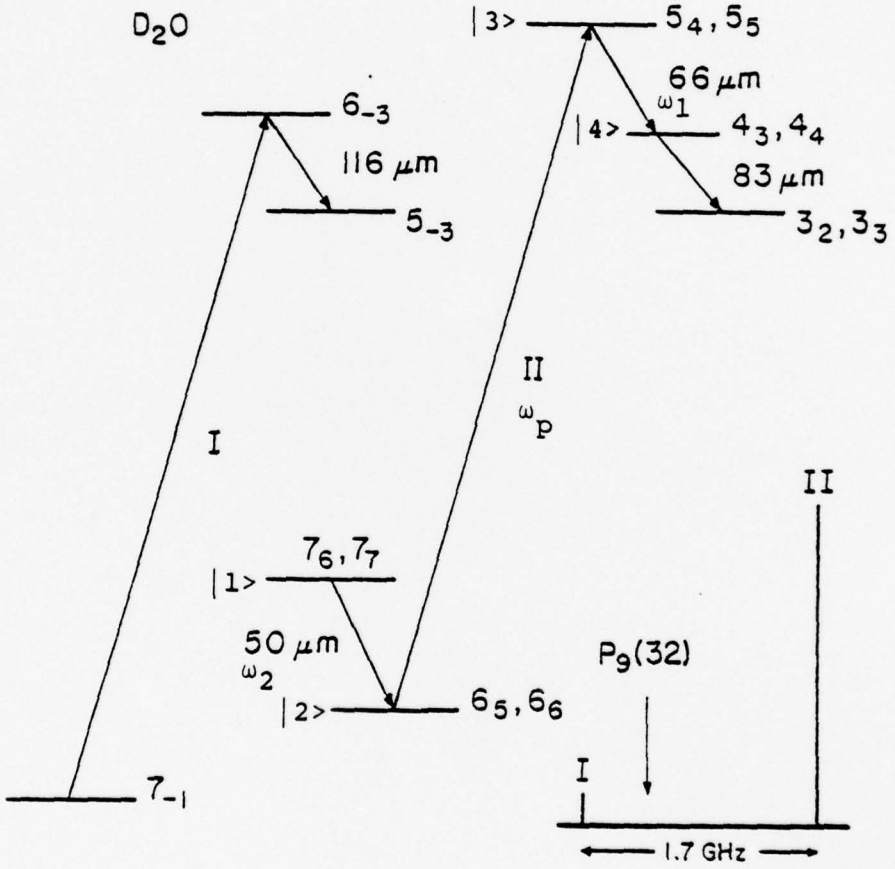

Fig. 12. Partial D_2O energy level diagram near the $P_9(32)$ laser line. Insert shows the detailed absorption spectra based on tunable diode laser spectroscopy of Ref. 13.

quasi-static density matrix formalism, and have derived exact algebraic expressions for wave gain profiles which point to nonlinear mechanisms of wave coupling. This analysis has been incorporated into a computer program in which the evolution of the 66 μm and 50 μm emission in a single-pass system is modeled.

B. Analysis

The four-level density matrix analysis applied to the D_2O system indicates that two-photon processes such as optically pumped lasing and stimulated Raman emission are significantly modified by an interaction with an intense third radiation field. The system of coupled density matrix equations has been solved quasi-statically (in the $t>>T_2$ limit), neglecting off-resonant and transient terms in favor of terms with small resonance denominators. Algebraically exact expressions have thus been derived for the off-diagonal elements of the density matrix in terms of arbitrary field intensities, population differences, detunings, and phenomenological dephasing and damping processes. Field gains can then be cast in a form in which terms are grouped by population differences and contributions to the net gain by one-,two-, and three-photon processes are thereby elucidated.

In the level configuration depicted in Fig.12, applicable to the D_2O system, the ω_1 gain assumes the form:

$$G_1 \propto \alpha_1(n_3-n_4) + \omega_p^2\,\alpha_2(n_2-n_4) + \omega_p^2\omega_b^2\alpha_3(n_1-n_4)$$

where (n_i-n_j) is the population difference between levels i and j, α_i are coupling factors peaked, respectively, near i-photon resonances, and ω_i are the Rabi frequencies. The α_i are field-dependent, and account for relative polarizations of the interacting fields as well as AC Stark shifts. Exact field intensity-dependent expressions have been derived for the AC Stark shifts and i-photon linewidths. An appropriate summation over sublevels incorporates the effect of the M-degeneracy of the rotational levels, lifted in the presence of intense optical fields.

In the D_2O system of Fig. 12, the 50 μm ground state transition cannot be inverted on a quasi-steady-state basis and laser loss is predicted on resonance. However a two-photon (SRE) gain multiplier, α_2, plotted in Fig. 13 as a function of frequency and 66 μm field intensity at a characteristic D_2O pressure and pump intensity, yields a net gain off-resonance under population conditions characteristic of a saturated 66 μm SRE process. This partially accounts for the observed delayed onset of the 50 μm signal (see Fig. 3 of Appendix I).

C. Experiment

The system on which our optically pumped D_2O research has been carried out is essentially that described in Appendix

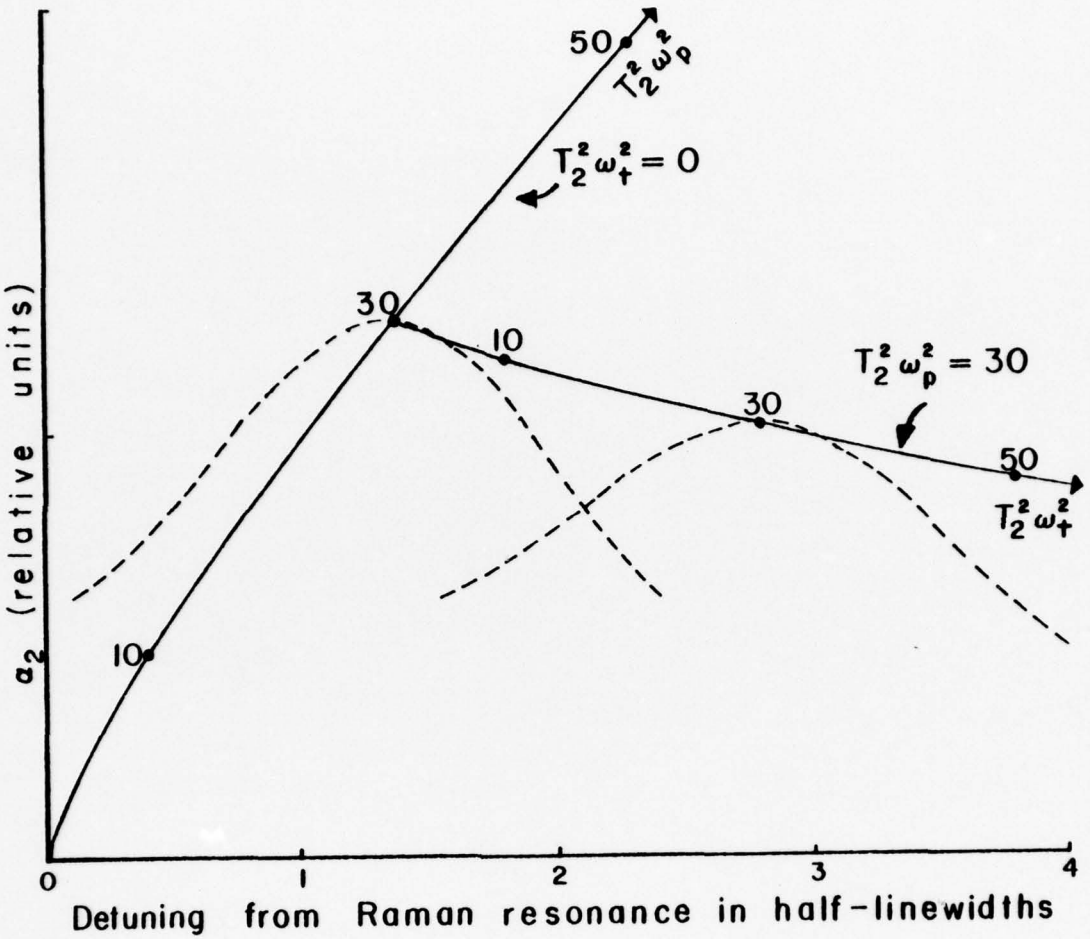

Fig. 13. Loci of maximum α_2 and detuning from Raman resonance at maximum α_2 pertaining to the 50 μm D_2O line, at a pump detuning of 1.1 GHz and a D_2O pressure of 2.2 torr. For a FWHM linewidth of 40 MHz/torr, $T_2^2\,\omega_p^2 = 50$ corresponds to a pump intensity of 822 kW/cm^2 while $T_2^2\,\omega_t^2 = 50$ corresponds to a 66 μm signal intensity of 2.9 kW/cm^2 . Two gain profile segments illustrate the line broadening influence of the 66 μm signal.

I. Detunings of pump and FIR fields were determined on the basis of pressure dependence of absorption in D_2O as described there. Subsequent modifications have included improvement of the TEA laser transverse beam quality and stability (by geometrical modifications of the CW amplifier section), and elimination of the Au-coated back reflector and all surfaces normal to the FIR beam to ensure single-pass operation. A cell containing cyclopropane at variable pressure in line with the pump beam allows continuous variation of pump intensity from shot to shot. Net 66 μm pulse energies and their shot-to-shot fluctuations have been measured as functions of cell length, pressure, and pump intensity in the single-pass configuration.

At a pump energy of 72 mJ/pulse, FIR energy fluctuations are characteristic of saturation of the 66 μm SRE process within a length $\sim$ 2.5 m, assuming an incoherent blackbody source at the input end of the cell.[15] Gain and threshold estimates derived from these data are reasonable within the context of our theoretical understanding of the system.

The propagation model entailed sampling the temporal shape of the pump pulse for boundary conditions for spatial integration of Dirac delta function pulses down the length of the cell. The viability of the model was established on the basis of a comparison of predicted with measured evolution of the waves, temporal relationships among the pulses, and pressure

dependence of peak intensities. The model fails, however, to account for observed 50 μm intensities at lower pressures, suggesting the possible significance of a backward-propagating 66 μm wave.

One further feature predicted by our analysis was also investigated. As the FIR gain profile is a function of field intensities, a chirp in the frequency of the emitted FIR pulse is to be expected as the pump pulse rises from FIR threshold to its peak value. This was studied by a time-resolved interferometric technique with the major goal of identifying the magnitude of the chirp and hence the 'start' condition for the emission. The absence of any observed effect is consistent with a predicted chirp in the 66 μm emission of $\sim$ 30 MHz at 4 torr.

D. Conclusions

Inroads have been made into the understanding of FIR emission by a molecular system optically pumped off-resonance in the infrared. The SRE mechanism whereby the strong 66 μm and the 50 μm lines are generated in D_2O has been identified for the first time. Research into the precise field and molecular population dynamics is well underway with implications for a variety of other configurations of coupled waves in gaseous systems.

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2. S. J. Petuchowski, H. Chung and T. A. DeTemple, "Diagrammatic Techniques for the Determination of AC Stark Shifts".

3. S. J. Petuchowski and T. A. DeTemple, "Multiphoton Interactions in Optically Pumped D_2O".

4. A. T. Rosenberger and T. A. DeTemple, "Far Infrared Superradiance in CH_3F".

V. SCIENTIFIC PERSONNEL ASSOCIATED WITH GRANT

1. Paul D. Coleman, Principal Investigator

2. Thomas A. DeTemple, Principal Investigator

3. Mark Gimple, M.S., EE 1976

4. Albert T. Rosenberger, Ph.D., Physics 1979

5. Samuel J. Petuchowski, Ph.D., Physics 1979

APPENDIX I.

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Stimulated Raman Emission in Infrared Excited Gases

S. J. PETUCHOWSKI, A. T. ROSENBERGER, AND THOMAS A. DeTEMPLE

Abstract—Using the pressure dependence of absorption, absorption coefficients and detunings were measured for CO_2 pump lines and the strong far infrared emission in optically pumped D_2O. The P (32) CO_2 line was found to be detuned ~1.5 GHz from the ν_2 band transitions $6_6, 6_5 \rightarrow 5_5, 5_4$. The resulting emission lines at 50.3 μm and 66 μm were found to be detuned from their respective transitions by about the same amount. On the basis of these measurements and gain estimates for the far infrared, the resulting emission lines are identified as stimulated Raman emission.

I. Introduction

INTEREST in far infrared (FIR) emission from optically pumped D_2O vapor stems from observations of high infrared to FIR conversion efficiency, ranking it along with $C^{12}H_3F$ as one of the stronger pulsed FIR sources [1]-[3]. Recent spectroscopic data have indicated that many of the pump lines are detuned many Doppler widths from their respective D_2O absorptions suggesting that off-resonant pumping is responsible for the strong emission [4]. Originally postulated to explain FIR emission in NH_3, off-resonant pumping is essentially wing absorption with resulting FIR emission on or near FIR line center [5]. An equally consistent and sometimes stronger off-resonant effect is stimulated Raman emission, which results in the FIR being emitted off-resonance by an amount equal to the pump detuning. In this article we present evidence for stimulated Raman emission in D_2O vapor.

In the next section, the experiment is discussed along with our recent spectral measurements and line identifications. In Section III, FIR frequency detuning measurements are presented along with Raman and laser gain estimates while the results are summarized in Section IV.

II. Experiment

In Fig. 1 is shown the experiment which was comprised of a CO_2 TEA laser, a 3.5 m long FIR cell, a grating monochromator, and an external absorption cell for wavelength and fine frequency measurements. The CO_2 laser operated on a single transverse and longitudinal mode, the latter obtained with the use of a CW low pressure CO_2 amplifier section. Using this technique the laser oscillated on CO_2 line center ±30 MHz with a single pulse spectral purity of better than 10 MHz including chirp [6].

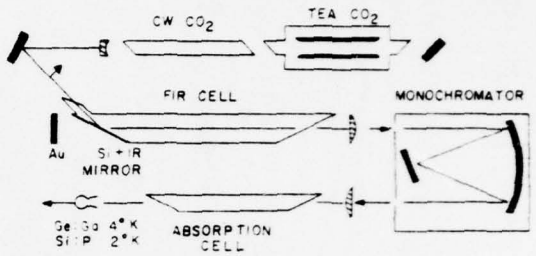

Fig. 1. Experimental arrangement. The absorption cell length was varied between 2 m and 20 cm.

The CO_2 pulse entered the FIR cell through a NaCl Brewster window and was reflected into the active region by a Si Brewster window. The interior portion of the Brewster window was coated with a multilayer Ge-ZnS IR mirror to provide >90 percent reflectivity throughout the CO_2 pump lines for the *P*-polarized pump [7]. This mirror had an estimated FIR absorption of <10 percent for wavelengths near 50 μm.

A single Au-coated flat was used as a back reflector while the output was transmitted either through another Si Brewster window or through a normal incidence high density polyethylene window. Most of the measurements were performed with the latter which implies a very low *Q* FIR cavity.

In view of the fact that there was only one line common to the previous two spectral measurements of the FIR emission from D_2O, we reanalyzed the spectral content associated with the strong emission using the P (32) 9.6 μm pump [1], [2]. This was performed with a $\frac{1}{2}$ m grating spectrometer using various FIR gratings in various orders and using higher orders of the weakly transmitted CO_2 as a wavelength marker. The accuracy of the measurements was ±0.1 μm with the results in basic agreement with previous observations [1].

Using the recent ν_2 band conventional spectroscopic measurements and resulting assignments of Shaw and Lin, we have been able to identify all IR and FIR transitions which are listed in Table I [4], [8]. The starred entries are new assignments. The notation is J_τ where $\tau = K_{-1} - K_{+1}$ and the strongly allowed transitions satisfy $\Delta J = 0, \pm 1$ and $\Delta\tau = 0, \pm 2$ [9].

A partial energy level diagram for the P (32) transition is shown in Fig. 2. Based on the results of Shaw and Lin, the insert shows the positions of the two transitions relative to

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TABLE I
D_2O ASSIGNMENTS

PUMP[a]	ABS. (ν_2 BAND)	FIR(μm)	TRANSITION	
P(32)	$^{*}6_{5,6} \leftarrow 5_{4,5}$	65.9μm	$^{*}4_{3,4} \leftarrow 5_{4,5}$	ν_2
		82.6μm	$^{*}3_{2,3} \leftarrow 4_{3,4}$	ν_2
		50.3μm	$^{*}6_{5,6} \leftarrow 7_{6,7}$	GND.
	$7_{-1} \leftarrow 6_{-3}$	119 μm	$5_{-3} \leftarrow 6_{-3}$	ν_2
R(12)	$^{*}10_{-8} \leftarrow 9_{-6}$	94 μm	$^{*}8_{-6} \leftarrow 9_{-6}$	ν_2
		114 μm	$^{*}9_{-8} \leftarrow 9_{-6}$	ν_2
		142 μm	$^{*}10_{-8} \leftarrow 10_{-6}$	GND.
R(22)	$5_0 \leftarrow 4_0$	385 μm	$4_{-2} \leftarrow 4_0$	ν_2
		358 μm	$4_{-4} \leftarrow 4_{-2}$	ν_2
R(32)	$9_{-9} \leftarrow 8_{-7}$	116 μm	$7_{-7} \leftarrow 8_{-7}$	ν_2
R(34)	$4_2 \leftarrow 3_0$	263 μm	$3_{-2} \leftarrow 3_0$	ν_2

[a] 9.6 μm bands
[*] Newly Assigned

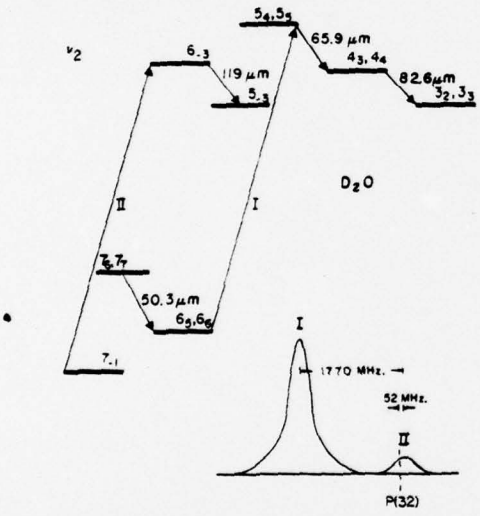

Fig. 2. Partial energy level diagram near the P (32) line. Insert shows the line positions and strengths based on conventional spectroscopic studies [4].

P (32) with the height being the relative absorption [4], [9]. The doublet transition from 6_5 and 6_6 is thought to be split by less than the Doppler width (50 MHz) [4]. Because the pump is essentially single frequency and detuned ~30 Doppler widths from the strongest absorption, the question arises as to the nature of the resulting FIR emission—laser or stimulated Raman. In what follows we present evidence that the 65.9 μm and the 50.3 μm transitions are due to stimulated Raman emission while the 82.6 μm appears to be a cascade laser transition. The 119 μm transition was not investigated.

Preliminary evidence for the Raman effect came from temporal measurements of the various signals shown in Fig. 3.

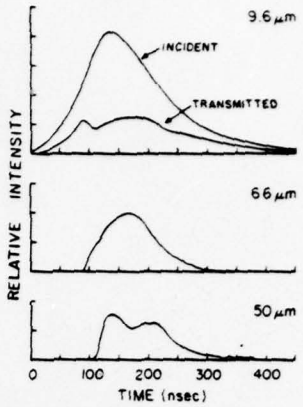

Fig. 3. Synchronized incident and transmitted CO_2 pulses, and emitted FIR pulses. Incident CO_2 ~ 0.6 MW, emitted FIR ~kW. The FIR detector was Si at 2 K with a speed of ~5 ns [10]. Source cell pressure: 3.4 torr.

Prior to the onset of the FIR, the CO_2 absorption coefficient was slightly less than the small signal value (to be discussed in the next section) indicating some small saturation. During the occurrence of the FIR, the peak absorption coefficient was a factor of 2 larger than the small signal value. This is incompatible with both FIR waves being on resonance because at best only the small signal absorption coefficient could be recovered by having saturating FIR waves present.

However, pump depletion and an apparent intensity dependent absorption coefficient are both characteristic of a strong parametric effect such as stimulated Raman emission. An alternate explanation of the increased absorption might be either a two-step or two-photon (IR + IR or IR + FIR) absorption. From the available spectroscopic data for ν_2, $2\nu_2$, ν_1, and ν_3, we have calculated that the smallest detuning for the latter processes is >10 GHz making the postulated Raman more favorable solely on the basis of detuning [4], [11].

III. Detuning Measurements

One of the major characteristics which distinguish the stimulated Raman signal from a laser signal is the optical frequency. In the latter case the frequency will be at the molecular frequency while in the former case, the frequency will be detuned from the molecular frequency by an amount equal to the pump detuning. In order to measure the expected small frequency shift, a second D_2O cell was used as a spectrometer, and is shown in Fig. 1.

For an assumed detuning much larger than the Doppler width, the wing absorption coefficient is given by

$$\alpha(\nu) = \frac{\lambda^2 A_{21}}{16\pi^2}\left(n_2 - \frac{g_2}{g_1}n_1\right)\frac{\Delta\nu_H}{(\nu-\nu_0)^2} \tag{1}$$

where 2 and 1 refer to the upper and lower levels, g_i is the level degeneracy, A_{21} is the reciprocal radiative lifetime, $\Delta\nu_H$ is the homogeneous line width (FWHM), and ν and ν_0 are the optical and molecular frequencies. Since both the population difference and the line width are proportional to pressure p, $\alpha \sim p^2$ with a slope inversely proportional to the square of

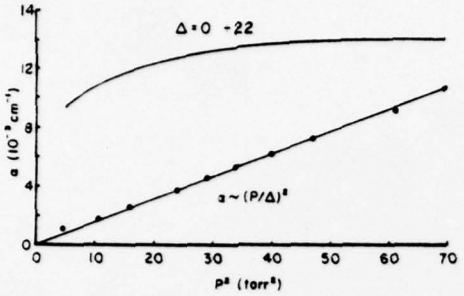

Fig. 4. Small signal absorption coefficient at P(32). Also shown is the line center absorption coefficient for I. Δ is the detuning from I.

the detuning. Hence, a measured value of α versus p^2 can yield the detuning [12].

There are two major sources of error involved in applying this technique to frequency measurements. The first is in the broadening rates for individual transitions. From available data in H_2O and D_2O, the broadening rates range from 26-60 MHz/torr implying a detuning error of ±25 percent for an assumed average broadening rate of 40 MHz/torr [2], [13], [14]. The second source of error is in the possibility of absorption due to nearby transitions, which is correctable only to within the accuracy with which the spectra are known. From available sources we estimate the accuracies to be: ground state FIR, ±100 MHz, ν_2 IR, ±400 MHz, ν_2 FIR ±400 MHz [15]-[17].

Because of the known uncertainties of the IR transitions, particularly II in Fig. 2, and as a check on the technique, absorption coefficients and detunings were measured for a few CW pump lines listed in Table I [18]. An example of the data for P(32) is shown in Fig. 4. The results are presented in Table II with the detunings obtained using $\Delta\nu_H = 40$ MHz/torr, a value based on our measured $R(22)$ absorption coefficient and the known detuning of -318 MHz [2], [12]. These data essentially confirm the magnitude of the calculated detunings [4].

For the case of P(32), the analysis was complicated by the presence of the two transitions shown in Fig. 2. For two absorbing transitions there are generally four possible line locations which result in the same value of absorption. For the specific case of widely separated strong and weak transitions, the locations are approximately symmetric about each transition. For the data in Fig. 4, the resulting candidate line positions are ±855 MHz about I and ±500 MHz about II. But because of the linearity of α in Fig. 4 and the variation in α over the tuning range of the CW CO_2 laser we have deduced that the most likely line location is $\lesssim$ -500 MHz from II and $\lesssim$ +1.5 GHz from I [19]. Hence, the pump appears to be above the I line center by ~30 Doppler widths.

The technique was then applied to the FIR signals. Prior to propagating through the absorption cell, the intense (~kW) FIR pulses were filtered and attenuated to low levels (<W) to prevent saturation. The apparent absorption coefficient was then measured as a function of pressure in both the source and absorption cells with the resulting data shown in Fig. 5. Each data point represents an *energy* absorption coefficient

TABLE II
MEASURED DETUNINGS

PUMP	α(/cm·torr²)	$(\nu-\nu_o)$CALC[a]	$\lvert\nu-\nu_o\rvert$MEAS[c]
P(32)	1.5×10^{-4}	+1.7 GHz(I)[b]	$\lesssim$ 1.5 GHz
		-52 MHz(II)[b]	$\gtrsim$ 500 MHz
R(12)	1.4×10^{-4}	836 MHz	680 MHz
R(22)	1.8×10^{-3}	-81 MHz	318 MHz[d]
R(32)	2.2×10^{-4}	773 MHz	670 MHz
R(34)	3.0×10^{-5}	-2.84 GHz	2.3 GHz

[a] From Ref. 4

[b] See Fig. 2

[c] Assuming $\Delta\nu_H$ = 40 MHz/torr

[d] From Ref. 2, also $\nu < \nu_o$ was determined

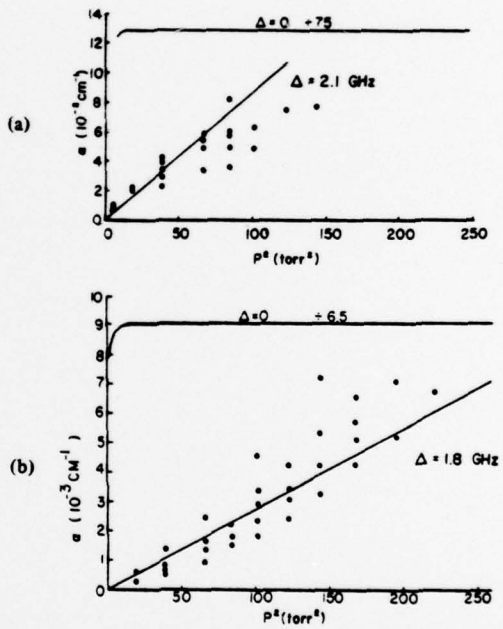

Fig. 5. (a) Measured FIR absorption coefficient for the 50 μm signal. Δ is the magnitude of the detuning from $6_{5,6} \rightarrow 7_{6,7}$. (b) Measured FIR absorption coefficient for the 66 μm signal. Δ is the magnitude of the detuning from ν_2 $4_{3,4} \rightarrow 5_{4,5}$. Also shown are line center absorption coefficients, and $\alpha(p^2)$ averaged over the experimental range of source cell pressures 1-5 torr.

averaged over ~100 FIR pulses. Part of the scatter is thus attributed to fluctuations in the pump and FIR amplitudes. As can be seen in this figure, the 50 μm signal appeared to be detuned 2.1 GHz, which is slightly greater than the estimated pump detuning of 1.5 GHz. The sign of the FIR detuning could not be determined.

For the 66 μm case, the absorption is dominated by the ground state transition $6_{-1} \rightarrow 7_1$ with a calculated location 5.75 GHz below the ν_2 FIR transitions. Because of the strong

ground state transition, there are only two candidate line locations, +1.8 GHz and -6.75 GHz relative to the 66 μm line center. The latter is rejected because it is inconsistent with any Raman or laser process. The weak 83 μm signal was absorbed strongly at all pressures and appeared to be on or very near laser resonance. A consistent interpretation of these results is that, by virtue of the measured IR and FIR detunings, both the 50 and 66 μm signals are due to separate stimulated Raman processes, with perhaps a small ac Stark shift, while the 83 μm signal appears to be a cascade laser transition. As a further check on this, we can estimate the respective small signal gain coefficients and Stark shifts.

Using a density matrix description of two waves interacting in a three-level system in the near-resonant approximation, the gain at the emitted frequency can be expressed as

$$G_f = \pm\sigma_{32}\left[F_1(n_3 - n_2) + F_2(n_1 - n_2)\right] \tag{2}$$

where the +(-) sign refers to the inverted (normal) vee configuration shown in Fig. 6 and σ_{32} is the homogeneous cross-section at line center for the $3 \rightarrow 2$ transition [20]. F_1 and F_2 are the laser and Raman cross section multipliers, respectively, with F_2 proportional to the pump intensity. The explicit forms of F_1 and F_2 are listed in the Appendix. In Fig. 6, F_1 and F_2 are shown versus the emitted frequency detuning for a fixed pump detuning with normalized pump intensity as a parameter. The values are representative of the experimental situation in Fig. 3. Two key features to note are that the laser and Raman resonances are ac Stark shifted by the same amount in opposite directions and that the magnitude of F_2 is not negligible compared with F_1. For the inverted vee configuration appropriate to the 66 μm transition, Raman gain exists for $n_1 > n_2$, which is certainly the case for weak pump saturation. For the normal vee configuration appropriate to the 50 μm transition, Raman gain requires $n_2 > n_1$, which is also satisfied for a weak pump [21], [22]. Hence, both transitions may undergo separate laser or Raman transitions.

The gain coefficients are estimated by approximating the populations with a steady-state value. For the conditions in Fig. 3 at the time of onset of the FIR, the Raman and laser gains at 50 μm are found to be 0.5 cm^{-1} and ~7 cm^{-1}, respectively (assuming 1.5 GHz pump detuning, $\beta = 0.1$ corresponding to ~300 kW/cm^2). For 66 μm the gains are estimated to be 0.7 cm^{-1} and 0.5 cm^{-1} for the Raman and laser cases. Because of the higher gain, the 66 μm Raman signal should build up first followed by the 50 μm signal. This is evident in Fig. 3.

Also evident in Fig. 3 is an inflection point in the 66 μm signal which correlates with the onset of the 50 μm signal. Considering the inverted vee configuration in Fig. 6, a 66 μm Raman process results in a preferential population of 2 with resulting wing absorption of the Raman signal due to the $2 \rightarrow 3$ transition. In contrast, a 50 μm Raman signal populates 3 which would decrease this absorption increasing the net 66 μm Raman gain and resulting output as observed.

It is also interesting to note the predicted loss for the 50 μm case. In fact even with a saturating pump and 66 μm signal, laser or Raman, there is still a predicted 50 μm laser loss of -0.22 cm^{-1}. In contrast, the 66 μm laser and Raman gains are very close, suggesting that other factors may influence the preferential growth of one. One such factor is the ground state absorption $6_{-1} \rightarrow 7_1$ which will be larger for the laser case than the Raman, favoring the growth of the latter.

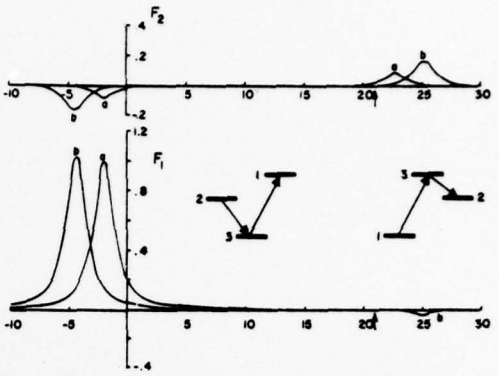

Fig. 6. Calculated values of F_1 and F_2 versus normalized FIR frequency $2(\nu_{32} - \nu_f)/\Delta\nu_H$. Labels a and b refer to β values of 0.1 and 0.3, respectively, with β proportional to pump intensity as defined in the Appendix. The arrows show the pump location. Insert shows two possible Raman configurations.

From Fig. 6, the ac Stark shift is estimated to be ~150 MHz assuming the detuning to be 1.5 GHz. Hence the Raman lines should be detuned at least 1.65 GHz whereas any laser lines would be detuned ~150 MHz. This is to be compared with the measured 66 μm detuning of 1.8 GHz and the 50 μm detuning of 2.1 GHz. Recalling the uncertainties in $\Delta\nu_H$, the agreement is reasonable and confirms the nature of the processes.

IV. Conclusions

Using the most recent high resolution spectral data, we have identified the strong FIR transitions associated with the P (32) pump. The expected pump detunings based on these data have also been quantitatively confirmed. Because of the large P (32) and emitted 50 and 66 μm signal detunings, the emission processes were identified as being due to two separate stimulated Raman effects. The expected small ac Stark shift of the FIR by the strong pump was not fully resolved. The major error in using the pressure dependence of the absorption for frequency measurements is in the uncertainties of the various broadening coefficients.

There are a number of interesting implications of these observations. First, a Raman process can be twice as efficient as a laser process because, in principle, every absorbed IR photon produces a Raman photon whereas laser emission is limited to only half of the excited molecules. Second, the strong-field near-resonant interaction produces an ac Stark shift which, because of the space-time variation of the pump, may chirp the FIR. In fact, the scatter evident in the data of Fig. 5 has been partially correlated with the source cell pressure and hence the FIR and IR intensities and their implied Stark shifts. Third, the existence of absorption by nearby ground state transitions may be a limiting factor in the overall FIR growth dynamics. For example, the ground state

transition $4_{-4} \rightarrow 4_{-2}$, with a calculated position 3.68 GHz above the 385 μm transition, has a line center absorption of 0.45 cm^{-1} which may be sufficiently strong to affect the FIR. Finally, because of the presence of two strong FIR waves, there may exist contributions to the dynamics associated with three-photon processes such as a double Raman or laser-Raman process.

It would appear that under the appropriate tuning conditions, many of the multiphoton processes so easily observed in the visible may also be observed in the IR and FIR. Recent observations of two-photon absorption (IR + IR) and IR Raman, and this observation of FIR Raman, suggest that, far from being weak, the multiphoton effects are quite strong, may already exist in a number of known off-resonant cases, and might be observed in three- and four-wave interactions [5], [23]-[25].

Appendix

For times longer than the inverse linewidth, an appropriate rate equation description of the inverted vee configuration in Fig. 6 is

$$\dot{n}_2 = -\gamma_2(n_2 - n_2^e) + G_f I_f$$

$$\dot{n}_1 = -\gamma_1(n_1 - n_1^e) + G_p I_p$$

$$\dot{n}_3 = -\gamma_3(n_3 - n_3^e) - G_p I_p - G_f I_f$$

where γ_i is the relaxation rate, n_i^e is the equilibrium population G is a gain coefficient, I_i the flux $\epsilon_0 E_i^2/2\hbar k_i$ with E_i the peak electric field and $k_i = 2\pi/\lambda_i$. The subscripts p and f refer to the pump and FIR. Using a density matrix description of a quasi steady-state two-wave interaction in the near resonant approximation (detunings are suboptical) and assuming all linewidths to be the same, the gain coefficients are found to be for the inverted vee case:

$$G_f = \sigma_{32}[(n_3 - n_2)F_1 + (n_1 - n_2)F_2]$$

$$G_p = \sigma_{31}[(n_1 - n_3)f_1 + (n_1 - n_2)f_2]$$

where σ_{ij} is the homogeneous cross section at line center [26]. Expressing detunings as $x = 2(\nu_{31} - \nu_p)/\Delta\nu_H$, $y = 2(\nu_{32} - \nu_f)/\Delta\nu_H$, normalized fields as $P = \mu_{13}E_p/h\Delta\nu_H$ and $S = \mu_{23}E_f/h\Delta\nu_H$ where μ_{ij} and ν_{ij} are the transition dipole moment and frequency; then defining $L(z) = z + i$, we find

$$F_1 = \mathrm{Im}\left(\left(1 + \frac{P^2 - S^2}{L(x-y)L(x)}\right)/L^*(y)\Delta\right)$$

$$F_2 = \mathrm{Im}\left(-P^2/L(x-y)L(x)L^*(y)\Delta\right)$$

$$f_1 = \mathrm{Im}\left(\left(1 + \frac{P^2 - S^2}{L(x-y)L^*(y)}\right)/L(x)\Delta\right)$$

$$f_2 = -\sigma_{32} I_s F_2/\sigma_{31} I_p$$

$$\Delta = 1 + P^2/L(x-y)L^*(y) - S^2/L(x-y)L(x).$$

These expressions contain ac Stark shifts due to *both* waves. In the weak field approximation $F_2 \simeq P^2/(x^2+1) = \beta$ at the Raman resonance ($y = x$). The data of Fig. 6 are for $S = 0$ and $\gamma_i = 2\pi\Delta\nu_H$.

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